

# **Exposure Analysis of Five Fish Consuming Populations for Overexposure to Methylmercury**

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# Executive Summary

## Introduction

Environmental exposure to human populations around the world from methylmercury has made this a chemical of global concern. Exposure through diet has resulted in increased body burdens of methylmercury in human populations. Prenatal exposure to the fetus can lead to central nervous system damage which can produce neurotoxic effects in children. Catastrophic exposure to communities in Japan and Iraq resulted in severe toxic and teratogenic effects. Results from these studies indicate that, with respect to exposure, the populations of greatest concern consist of women of child bearing age and their offspring. Whether low-level methylmercury exposures will produce delayed effects in these populations or if the effects observed in the cohorts are chronic or transient remains to be determined. There is no longer doubt about the ability of methylmercury to produce deleterious effects in animals and humans.

In Washington State various subsistence and recreational fishing populations consume fish species that contain methylmercury. The goal of this study was to determine if various populations in this state are consuming contaminated fish in quantities that could possibly result in deleterious outcomes. To accomplish this, consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species were examined. These populations are: recreational anglers consuming fish from Lake Roosevelt, recreational shore and boat anglers consuming fish from Puget Sound, the Tulalip Tribes, the Squaxin Island tribe, and the Suquamish tribe. The consumption rates for these populations were used with fish contamination data to analyze exposure to methylmercury. Concomitantly, a Tolerable Daily Intake (TDI) was derived for methylmercury (0.035 to 0.08 µg/kg/day). The TDI is unlikely to result in adverse health effects and is based on scientific studies investigating sensitive endpoints in children of mothers who consume fish over prolonged periods of time. A more extensive discussion of this TDI can be found in a previous report produced by this office. The results from the exposure analyses for the diverse populations were compared with the TDI to determine if certain populations were exceeding this health effects value.

## Results

### *Recreational anglers consuming fish from Lake Roosevelt*

Angler consumption patterns were determined and used with mercury fish tissue contamination data to determine if this Lake Roosevelt fish consuming population is exposed to levels of mercury that are above levels deemed to not result in adverse health effects. Results from the exposure analysis indicate that, at a minimum, 87% of the anglers surveyed have intake levels below the upper bound of the TDI. Individuals exceeding the TDI were those who ate fish more frequently and in greater quantity than the average angler. Nearly all individuals exceeding the TDI were adult males estimated to be greater than fifty years of age. If the spouses of these older adult males are of approximately the same age, they also

would not represent a population of concern (women of child bearing age). Based on the age structure of the individuals interviewed, there does not appear to be a population consisting of women of child bearing age consuming fish resulting in intake levels above the TDI. As the benefits of consuming fish as a source of protein compared to other protein sources is well documented, suggesting that older adults, male or female, consume less fish may have a deleterious impact. However, to reduce exposure to mercury in individuals that eat fish more frequently and in greater quantity, educational efforts informing them that consuming a variety of fish species from a variety of locations could be of benefit.

### ***Recreational shore and boat anglers consuming fish from Puget Sound***

The exposure analyses for the recreational shore and boat anglers suggests that mercury intake levels are below the TDI. However, anglers consuming chinook salmon from one particular location (Elliot Bay) have methylmercury exposures (0.09 µg/kg/day) just slightly exceeding the TDI. This result is significant for two reasons. First, concentrations of methylmercury are considered to be uniform for this species in Puget Sound suggesting that anglers who consume greater than 0.8 g/kg/day of chinook salmon would be exceeding the TDI. Second, the result of this exposure analysis was based on a geometric mean value, with no distribution data being available, indicating that individuals consuming above this value are also exceeding the TDI. Further consumption pattern data will be required to determine if this historical consumption data is still valid and if recreational anglers who represent our population of concern are exposed to mercury levels above the TDI. Presently, determining a clear course of action is difficult. Evidence does not exist to warrant a recommendation that recreational anglers who frequently consume chinook salmon should reduce their consumption rates, however, this is an issue definitely requiring further evaluation.

### ***Tulalip and Squaxin Island Tribes***

Several conclusions can be derived from the Tulalip Tribes and Squaxin Island tribe data. The 90<sup>th</sup> percentile total finfish consumption values for both tribal groups are approximately equal to the sum of 90<sup>th</sup> percentile values for each of the fish categories (anadromous, pelagic, bottom), suggesting that individuals consuming elevated amounts of fish from one category may be eating elevated quantities from the other two also. Although the contribution of mercury intake from pelagic and bottom fish may be small compared to anadromous fish, the results suggest that at least 8 – 14% and 10 – 25% of the Tulalip and Squaxin Island tribal members, respectively, have mercury intake levels above the TDI. Clearly, individuals within both populations are exceeding the TDI, however, the percent population value of 8 – 14% for the Tulalip Tribes that may have intake levels above the TDI will require validation through consumption distribution data.

Given the cultural, spiritual, and historical significance of fish consumption by tribal members, the suggestion of dietary changes may produce no clear benefit and could even result in deleterious health effects. Recommending changes must be weighed against the benefit of cultural events such as tribal ceremonies which are attended frequently and are a significant and important sources of fish. Present consumption data indicate that the mercury intake range in Tulalip Tribes (using the 86<sup>th</sup> and 90<sup>th</sup> percentiles) is 0.07 – 0.2 µg/kg/day, with the latter value based on the assumption that all individuals consume only chinook salmon. Until improved consumption data are available to better determine the percent of the

population exceeding the TDI, most if not all individuals within this population could achieve mercury intake levels at or below to the TDI while maintaining their cultural heritage by consuming other anadromous fish as alternatives to chinook.

The data for the Squaxin Island tribe present a much different dilemma. Results from the exposure analysis indicates that many individuals (25% of the Squaxin Island tribe) are consuming anadromous fish in quantities that may result in a mercury intake above the TDI. This conclusion is based on the assumption that chinook salmon are the only anadromous fish consumed. The consumption of other salmonids (i.e. chum, coho, steelhead, sockeye and pink) could reduce this value to approximately 10% of the population. Using the 90<sup>th</sup> percentile values also reflects that individuals have intake levels above the TDI. At a minimum, women of child bearing age within this population should be encouraged through educational efforts to consume salmonids other than chinook. As changing consumption of salmonid species will still result in intake levels above the TDI, the potential for adverse outcome remains. Simply suggesting that fish be consumed in lesser quantities is not, however, necessarily a prudent public health recommendation. Recommending a change in diet away from nutritional foods such as fish does not assure that the replacement will be equally beneficial. Also, as stated, cultural, spiritual, and historical practices must be considered. Educational efforts could be provided to encourage the broadening of fish species consumed to include freshwater fish species or pelagic fish that possibly have lower mercury concentrations (e.g. cod, pollock, herring, perch). Also, as hair analysis is presently the exposure metric most frequently relied upon, hair levels of methylmercury in the women of childbearing age who consume fish in excess of 1.0 g/kg/day should be monitored. These data could be used along with educational efforts as excellent tools for properly protecting the health of this population.

### ***The Suquamish Tribe***

The Suquamish tribe data suggest that individuals within the population have mercury intakes above the TDI. As with the above tribes, the 90<sup>th</sup> percentile total finfish consumption values are approximately equal to the sum of 90<sup>th</sup> percentile values for each of the fish categories suggesting that individuals consuming elevated amounts of fish from one category may be eating elevated quantities from the other two. The mercury intake range for the 90<sup>th</sup> percentile of these 92 individuals was estimated to be (0.16 to 0.34 µg/kg/day) with approximately 25% of those interviewed having mercury intake levels above the TDI. However, factors suggest that the actual mercury intake levels may be nearer the lower end of this range. Even considering the lower end of the range established from the exposure analysis (0.16 µg/kg/day), the TDI is still being exceeded by two-fold. The 75<sup>th</sup> percentile consumption rate yields an intake equivalent to the upper bound value of the TDI. Thus, 10% of the surveyed population sampled exceed the TDI by two-fold while 25% are at or above the TDI.

Inherently, this data set has similar difficulties to those observed with the Tulalip Tribes data as it is not known if 92 individual respondents are a representative sample. However, until improved consumption distribution data become available, women of child bearing age within this population should be encouraged through educational efforts to consume salmonids other than chinook. Many individuals having intake levels above the TDI could



reduce mercury intake levels, while maintaining their cultural heritage, by minimizing chinook salmon consumption. Rockfish consumption needs to be decreased by those consuming elevated quantities in this population. Eight of the ten individuals consuming rockfish, consume at the rate of approximately 0.124 g/kg/day (75<sup>th</sup> percentile) or less. Assuming Quilback and Copper rockfish are eaten equally, this results in a mercury intake level of 0.03 µg/kg/day or less, which is well below the TDI. However, those two individuals responsible for the 90<sup>th</sup> percentile consumption value of 0.789 g/kg/day have mercury intake levels of 0.18 µg/kg/day. As the rockfish contaminant level data did not come from the specific fishing areas used by this population, rockfish that this population regularly consume should be analyzed for mercury so that an accurate determination can be made of how much is being consumed. Many of the same recommendations made with respect to the other tribes applicable here, including consumption of other fish species having lower mercury concentrations and monitoring hair levels of methylmercury in the women of childbearing age who consume fish in excess of 1.0 g/kg/day as a preventive health measure.

## **Conclusion**

We examined consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species. The consumption rates were used with fish contamination data to analyze exposure to methylmercury and to determine if any of these populations exceeded a derived TDI for methylmercury (0.035 to 0.08 µg/kg/day). Results from the exposure analyses indicate that recreational anglers consuming freshwater species have exposure levels below the TDI as do recreational anglers consuming saltwater fish species with the exception of those consuming chinook salmon from one particular location (industrialized). The exposure analyses also indicate that many within the Native American populations exceed the TDI and that this occurs even though the mercury concentrations in certain fish species are comparable to concentrations found in fish from open waters where “background” levels are assumed. As with any TDI, changes to the value must be made as further data become available. Without maintaining diligence in this area, the result could be public health protection through a TDI that is too high which allows for individuals to be exposed to deleterious levels or through a TDI that is too low which would provide for a large gap between acceptable intake levels and those levels that cause toxic effects. With respect to mercury, it is imperative that this gap be minimized as a TDI that is too low will be a public health detriment as it results in recommendations that restrict or alter the consumption patterns of healthy food sources.

Also of import are “background” mercury levels present in salmon resulting in populations being exposed to mercury levels above the TDI. Regardless of whether the levels are “background” or above, public health protection can only be achieved by ensuring that exposure levels remain within present protective and accepted values, even if this impacts the consumption patterns of a particular fish type (chinook salmon) which is otherwise considered a very healthy food source. This is, however, not to suggest that other protein sources beside fish should be considered, but only that a variety of fish be consumed as the benefits of this protein source compared to others is well documented. In the case of Washington State, we must ensure that various salmon species are available for consumption so that chinook salmon are consumed by choice and not necessity.

The approaches used to protect individuals require difficult decisions when cultural and historical considerations must be considered or when intake levels just slightly exceed a value or set of values deemed to be protective. In this study, recommendations for how best to serve the public health needs of these populations regarding methylmercury exposure have been provided. The study outcomes presented herein allow state health departments and local health jurisdictions to develop intervention and education strategies to protect individuals, especially women of child-bearing age, from overexposure to methylmercury.

## **Exposure Analysis of Five Fish Consuming Populations for Overexposure to Methylmercury**

### **Abstract**

Mercury, in the form of methylmercury, is found in a myriad of fish species consumed by recreational and subsistence fishers around the world. Many agencies have attempted to address the issue of mercury consumption, resulting at times in the placement of advisories on waterways used for fishing. In this study, consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species were examined. These consumption rates were used with fish contamination data to assess the level of exposure to methylmercury and to determine if any of these populations exceed a derived tolerable daily intake (TDI) for methylmercury (0.035 to 0.08  $\mu\text{g/kg/day}$ ). The TDI is unlikely to result in adverse health effects and is based on scientific studies investigating sensitive endpoints in children of mothers who consume fish over prolonged periods of time. Results from the exposure analysis indicate that many within the Native American populations exceed the tolerable daily intake. This occurs even though the mercury concentrations in certain fish species are comparable to concentrations found in fish from open waters where “background” levels are assumed. Recreational anglers consuming freshwater species have exposure levels below the TDI as do anglers consuming saltwater species with the exception of those consuming fish from one particular location. Similar populations or populations having comparable consumption patterns of fish with equal or higher mercury levels in other areas will also exceed the tolerable daily intake level for mercury. The public health implications of this exposure analysis are discussed.

## **Introduction**

Environmental exposure of human populations around the world to methylmercury has made this a chemical of global concern. Exposure through diet has resulted in increased body burdens of methylmercury in human populations (ATSDR, 1997; USEPA, 1997).

Catastrophic exposure to communities in Japan and Iraq resulted in severe toxic and teratogenic effects (Harada, 1995). Laboratory studies in animals, both rodent and non-human primate, have revealed various reproductive, developmental and neurological effects from exposure (Khera, 1973; Chang et al., 1974; Sato and Ikuta, 1975; Bornhausen et al., 1980; Mohamed et al., 1987; Burbacher et al., 1988; Mitsumori et al., 1990). Methylmercury can readily cross the placental barrier following adult exposure (ATSDR, 1997; USEPA, 1997). Prenatal exposure to the fetus can lead to central nervous system damage which can produce neurotoxic effects in children (ATSDR, 1997; USEPA, 1997). These findings have led to the developing nervous system being considered the most sensitive endpoint (ATSDR, 1997; USEPA, 1997). Effects observed following in utero exposure in poisoning episodes have included blindness, deafness, abnormal reflexes, impaired motor development, spasticity, seizures and deficiencies in memory, learning and psychological parameters (ATSDR, 1997). Three major scientific studies that have provided great insight into the possible effects from exposure to methylmercury were recently conducted in New Zealand, the Seychelles Islands and in the Faroe Islands (Davidson et al., 1995, 1998; Grandjean et al., 1992, 1994, 1995, 1997, 1998; Kjellstrom et al., 1986, 1989; Marsh et al., 1995; Myers et al., 1995). Results from these studies indicate that, with respect to exposure, the populations of greatest concern consist of women of child bearing age and their offspring. Whether low-

level methylmercury exposures will produce delayed effects in these populations or if the effects observed in the cohorts are chronic or transient remains to be determined.

While there is no longer doubt about the ability of methylmercury to produce deleterious effects in animals and humans, determining the exposure level that will not cause concern for the health of offspring of mothers exposed for a finite period of time is problematic. This difficulty stems from the fact that while developmental and neurotoxic effects of methylmercury at elevated levels of exposure have been well-studied, the effects from low levels of exposure and from in utero exposures are only now becoming understood.

In Washington state various subsistence and recreational fishing populations consume fish species that contain methylmercury. The goal of this study was to determine if these populations are consuming contaminated fish in quantities that could possibly result in deleterious outcomes. To accomplish this, consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species were examined. The consumption rates were used with fish contamination data to analyze exposure to methylmercury. To establish if exposure could possibly result in deleterious outcomes, a tolerable daily intake (TDI) for methylmercury that is unlikely to result in adverse health effects was derived. The results from the exposure analysis are compared with the TDI and the public health implications of the results are discussed. Similar populations or populations having comparable consumption patterns of fish with equal or higher mercury levels in other areas will also require similar attention. The type of approach presented yields results that allow state health departments, local health jurisdictions and various agencies to develop intervention and education strategies to protect populations from overexposure to methylmercury.

## **Methods**

### **Consumption by recreational anglers (freshwater fish species)**

A fish consumption survey was conducted at Franklin D. Roosevelt Lake (Lake Roosevelt) where chemical contaminants have been measured in fish. This lake was chosen for this study because it: is visited by over one million people annually, is part of the Coulee Dam National Recreation Area, contains various fish species contaminated with methylmercury, and is bordered by both the Colville Indian and Spokane Indian Reservations. Lake Roosevelt, located in the northeast corner of Washington State, was formed in 1941 by the completion of Grand Coulee Dam on the Columbia River and extends 151 miles to within 15 miles of the United States/Canadian border. The study was conducted during 1994 and 1995 to determine the consumption patterns of anglers who repeatedly fish the lake with the presumption that individuals who repeatedly fish the lake, catch and consume the greatest amount of fish (DOH, 1997).

Fish consumption data from Lake Roosevelt anglers were collected by the Spokane Tribe of Indians in concert with on-going creel data collection for the Lake Roosevelt Monitoring Program. Lake Roosevelt was divided into three sections; upper, middle, and lower, with one creel clerk assigned to each section. Morning and afternoon survey locations were randomly selected from a total of 48 possible locations (Underwood et al., 1996a and b). The systematic sampling scheme consisted mainly of interviewing a single angler per boat upon return from their fishing trip, primarily at National Park Service boat launch facilities. The majority of respondents were boating anglers; there are few shore anglers in Lake Roosevelt. Due to length-biased sampling, anglers who frequent the lake most had the greatest likelihood of being surveyed. Fish consumption data were collected on a

questionnaire that was separate from the creel survey questionnaire. Creel data were collected first and included trip length information, fish species, length, and weight (Underwood et al., 1996a and b). Due to logistical limitations, creel clerks were instructed to collect fish consumption data from only one member of a fishing party, unless time allowed and members could be spatially separated to minimize question response bias. The consumption survey was brief so as not to overly burden anglers in light of the combined fish consumption and creel data collection efforts. Sex, age, and race information provided on the consumption survey reflected surveyor judgment, and were not the result of explicit questioning.

Interviews were conducted from August through November of 1994 and from May through September of 1995. These periods were selected based on high angler catch and pressure data for frequently caught Lake Roosevelt fish species (Griffith and Scholz, 1991; Thatcher et al., 1993; Thatcher et al., 1994). Interviews conducted in 1994 were limited to the middle and lower section of Lake Roosevelt whereas interviews conducted in 1995 included all three sections of Lake Roosevelt. Creel clerks familiar with field survey data collection methods conducted the interviews and were instructed specifically on this questionnaire prior to study initiation. A pilot survey was conducted to aid in questionnaire design.

A total of 448 interviews were conducted (231 in 1994, 217 in 1995). Data from anglers who did not consume Lake Roosevelt fish (10 in 1994, 4 in 1995), along with data from anglers who were previously surveyed (19 in 1994, 38 in 1995) were excluded from analysis. This resulted in 377 surveys for use in assessing fish consumption of the target population.

### **Consumption by recreational anglers (saltwater fish species)**

Information on saltwater fish consumption practices of anglers was obtained from a two-year study conducted in 1985 and 1987 in which 4,181 individuals, predominantly shoreside anglers, were interviewed in three locations in Puget Sound, WA (Landolt et al., 1985). In the second year of the study 437 individuals, predominantly boating anglers, were interviewed in two of the locations from the first year's study (Landolt et al., 1987).

Objectives of this two year study were to determine consumption rates of fish, to demographically characterize the population of anglers fishing in urban embayments, and to determine contaminant levels in edible portions of the most commonly caught fish species. Populations identified included Caucasian, Black, Hispanic, American Indian, and Asian (Korean, Phillipino and Southeast Asian). Fish species consumed by these populations included squid, hake, tomcod, pollock, salmon, flounder, perch, sole and rockfish.

### **Consumption by Tulalip and Squaxin Island Tribes**

Fish consumption rates were also obtained from a survey of the Tulalip and Squaxin Island Tribes of the Puget Sound Region (Toy et al., 1996). In 1994, 190 adults were interviewed (72 and 117 from Tulalip and Squaxin Island Tribes, respectively) to determine fish and shellfish consumption rates. Data were collected on species consumed, fish parts consumed, preparation methods, sources of fish, and children consumption rates.

Consumption rates were weight-adjusted and stratified based on tribe, age, gender, income and fish species group. Fish species consumed were grouped as anadromous, bottom, pelagic and shellfish with anadromous fish and shellfish being most frequently consumed.

### **Consumption by Suquamish Indian Tribe**



Also incorporated are the study results from a recently completed survey of the Suquamish Indian Tribe of the Port Madison Indian Reservation in Puget Sound. This study consisted of interviewing 92 adult tribal members during March of 1997 (The Suquamish Tribe, 1999). The objectives for this study were to ascertain seafood consumption rates, patterns and habits of members of the tribe. Person-to-person interviews were given and the survey was based on a stratified random sampling design. Fish species consumed include types of salmon/steelhead, halibut, pollock, perch, greenling sole, flounder, rockfish, sturgeon, tuna etc. Data were aggregated into groups for comparison with the Toy et al. 1996 results.

### **Contamination Data**

Mercury concentrations in fish and shellfish were obtained for various waterbodies from existing data sets (Landolt et al., 1985; Landolt et al., 1987; Johnson et al., 1988; Norecol, 1989; Serdar et al., 1993; Munn et al., 1995; West and O'Neill, 1995; PSWQA, 1995; Munn and Short, 1997; PSWQAT, 1998; O'Neill et al., 1998). The mercury concentrations used in this analysis came from the fishery that each specific population was using. Fish tissue mercury concentration data were compiled and are discussed in the Results section.

## **Results**

### **Tolerable Daily Intake**

Considerable effort has been put forth by various agencies, groups, and individuals to determine a mercury exposure level that would not result in adverse human health effects. Target-organ dose-based dose-response relationships would be instrumental in determining the levels of *in utero* methylmercury exposure that lead to neurological effects in infants and

children; however, available human and animal data do not presently allow for adequate relationships to be derived that can be used to protect public health. In lieu of this, other approaches have been used to determine acceptable or tolerable mercury intake levels.

Conditions of acute methylmercury exposure have shown the fetus to be particularly sensitive to methylmercury with adverse effects on infant development being documented in the absence of maternal toxicity or clinical illness (ATSDR, 1997; USEPA, 1997). Stern (1993) considered the available human and animal study data addressing developmental endpoints and suggested that the weight of evidence indicated the Reference Dose (RfD) to be 0.07  $\mu\text{g/kg/day}$ . With the understanding that the fetus and infant are more sensitive to adverse effects from methylmercury exposure, Gilbert and Grant-Webster (1995) used the Iraq episode data (Amin-Zaki et al., 1976, 1979, 1981; Bakir et al., 1973., Marsh et al., 1981, 1987), supported by data on neurobehavioral effects in animals, to develop an RfD range of 0.025 to 0.06  $\mu\text{g/kg/day}$ . Zelikoff and co-authors (1995) considered various approaches for establishing an RfD based on prenatal methylmercury exposure effects in small mammals, nonhuman primates and humans. These approaches are provided since establishing an RfD based on adult clinical effects may not be appropriate for protecting the developing fetus. The lowest RfD, 0.01  $\mu\text{g/kg/day}$ , was derived from effects seen in rats where abnormal performance in behavior was observed using a differential reinforcement of high rates paradigm. Rice (1996) has suggested that RfDs derived from animals are in agreement with those obtained from human data. By applying methods used by USEPA, Macaque monkey data from two data sets using different exposure regimens, but identical daily dosages (50  $\mu\text{g/kg/day}$  methylmercury) yielded an RfD of 0.05  $\mu\text{g/kg/day}$  (Gilbert et al., 1993; Rice, 1992). Monkeys dosed at 10 or 25  $\mu\text{g/kg/day}$  *in utero* until age four showed developmental

effects due to sensory-system impairment (Rice, 1992, 1996). These data lead to an RfD of 0.01  $\mu\text{g/kg/day}$ . The recently released Mercury Report to Congress (1997) by the USEPA provided an RfD for methylmercury, 0.1  $\mu\text{g/kg/day}$ , based on bench mark dose (BMD) modeling using the results from the study on Iraqi mother-child pairs. The Seychelles Islands study, the Faroe Islands study and other recent studies were not included in deriving this RfD. ATSDR (1997) has used the median hair level of the entire Seychelles Islands study cohort, with uncertainty factors, to derive a Minimal Risk Level for methylmercury of 0.3  $\mu\text{g/kg/day}$ . Clewell and co-workers (1998) used the results from several child development tests obtained from the Seychelles study to derive a Bench Mark Dose (BMD) in hair of 20 ppm methylmercury. This BMD along with daily ingestion rates allowed for the determination of RfDs that range from 0.3 to 1.0  $\mu\text{g/kg/day}$  (median 0.54  $\mu\text{g/kg/day}$ ).

The most common route of exposure to methylmercury is through fish consumption, with the exposure period of concern being long-term and with sensitive endpoints being impaired neurological development and long-term and/or delayed sequelae in children of exposed mothers. As a result, studies investigating sensitive endpoints in children of mothers consuming fish over prolonged periods of time are preferable to rodent and non-human primate data or to human data addressing endpoints where exposure was through another means or for shorter time periods. Presently, the investigations completed or ongoing in New Zealand, the Seychelles Islands and the Faroe Islands meet these criteria. These are prospective longitudinal studies of cohorts in which attempts were made to control for many of the factors that could influence child development, resulting in increased ability to detect neurological effects and delayed sequelae from methylmercury exposure.

The Seychelles Islands study investigated the effects of prenatal exposure to methylmercury through maternal fish consumption by testing for fetal neurodevelopmental effects at several timepoints during infancy and childhood. The main cohort consisted of 740 mother-infant pairs with no definite adverse neurodevelopmental effects observed in the offspring from low-level methylmercury exposure. The Faroe Islands cohort was generated from 1022 consecutive singleton births during 1986 and 1987. The cohort of children born to mothers with an average mercury hair level of 4.3 ppm (n = 917) underwent neurobehavioral examination at age seven. Although no mercury-related abnormalities were observed based on clinical examinations, mercury-related neuropsychological dysfunctions in language, attention and memory were observed. Whether these low-level methylmercury exposures will produce delayed effects in these populations or if the effects observed in the Faroe Islands cohort are chronic or transient remains to be determined. The New Zealand study investigated the effects on children from prenatal exposure to methylmercury in a population that consumed methylmercury contaminated fish (Kjellstrom et al., 1986, 1989). The cohort was made up of 237 6- and 7- year old children. As with the Seychellois population study, this study relied on continuous scale evaluations of cognitive function and on evaluations of subclinical neurological developmental performance. Results suggested developmental effects had occurred in children whose mothers had methylmercury hair levels of 6 ppm and above. A re-evaluation of these data using benchmark dose (BMD) modeling resulted in a statistical lower bound maternal hair mercury level, which can be used to determine acceptable human exposures, that ranges from 7.4 - 10 ppm (Crump et al., 1998).

Clewell and co-workers (1998) and ATSDR (1997) have considered the available data and used the Seychelles data and corroborating evidence from the studies in New

Zealand and Faroe Islands to derive RfDs. Contrary to this approach, the USEPA's Science Advisory Board has recommended that no changes to USEPA's Iraqi study-based RfD be made in the Mercury Report to Congress until further and more definitive data from the Seychelles Islands and Faroe Islands become available.

These discrepancies in approaches used to determine tolerable daily intake levels or reference doses, which include different approaches to address uncertainty, have occurred for many reasons dealing primarily with the limitations of the available data sets from the various studies. For example, the Iraqi population is considered not to be representative of a sensitive subpopulation within the perinatal group when compared to the populations from the Faroe Islands, the Seychelles Islands, and New Zealand (Cicmanec, 1996). Exposures to the mothers in the Iraqi episode were short term and at much higher levels than those exposures resulting from fish consumption. With respect to the Seychelles Islands, seafood contains essential nutrients, such as n-3 fatty acids, that could act as effect modifiers for methylmercury whereas in the Faroe Islands, these nutrients as well as PCBs could be effect modifiers. Grandjean and co-workers (1998) have suggested though that there is no clear association between PCB exposure and neuropsychological test results in children from the Faroe Islands. Also, as the fatty acid intake is reasonably high in both populations, it is possible that they may affect both populations in the same way, and would, therefore not be responsible for the differences in how methylmercury acted between these two populations. Although not observed in the Faroe Islands cohort, the Seychelles Islands cohort is achieving some developmental milestones more quickly than children in western cultures (Davidson et al., 1998). However, the correlation coefficient values for the regression analyses of the full model for many of the endpoints studied in the Seychellois population were low, indicating

that it was not possible to include all consequential factors (i.e. genetic variability or predisposition) that predict infant development in the model and, as a result, the tests conducted may lack the sensitivity to detect subtle developmental effects (Davidson et al., 1995, 1998). This lack in sensitivity, resulting in low correlation coefficients, may be more applicable to the early assessments conducted at the ages of 6 months and 19 months than those conducted at a later age (Davidson et al., 1995, 1998). The authors suggest that the number of individuals exposed at levels where effects appear may be too small to have a great deal of confidence in the results (Davidson et al., 1995, 1998). Also, there are potentially many population-specific considerations and confounding factors which make the basis for the rapid achievement of these developmental milestones unclear.

Based on the present results, various individuals and organizations have taken different approaches to deriving safe or tolerable consumption levels for human populations. The USEPA levels continue to be based on data from Iraq, while awaiting more conclusive results from the Seychelles Islands and Faroe Islands. Others have decided that the current data from the Seychelles Islands can be used at this time. Since public health protection is typically required without complete scientific clarity, it is clear that the process of establishing tolerable consumption levels will be ever evolving and responding to new findings as they become available. Whereas present available human and animal data are sufficient to attempt public health protection, great strides in determining these protective levels will be made as the *in utero* exposure knowledge base is further delineated. An exposure level that will render a platform from which to protect public health, however, can not be provided without giving foremost consideration to those studies suggesting that effects may occur at low levels of exposure. Preferentially relying on data indicating that no

observable deleterious effects arise from chronic low level mercury intake, as is the case with the Seychellois population data, would undermine public health principles which necessitate that observations indicating causal effects from exposure require consideration when protecting public health. As a result, the findings from the New Zealand study and the available findings from the Faroe Islands study must be considered even though significant work from the Seychelles Islands may provide a contrasting view.

Despite the respective shortcomings of the Faroe Islands, Seychelles Islands and New Zealand studies (e.g. lack of dietary information for cohorts in the studies), all are based on sound scientific foundations. While the Seychelles Islands data indicate a 10 ppm maternal hair mercury level to be associated with no deleterious effects, the New Zealand study suggests that maternal hair mercury levels of 6 ppm and above may be associated with brain function alterations in offspring (Kjellstrom et al., 1986; Davidson et al., 1998). A re-evaluation of these data, relying on a benchmark dose calculation, resulted in a maternal hair mercury level that ranges from 7.4 - 10 ppm (Crump et al., 1998). The authors of the Faroe Islands studies indicate that early dysfunction in children is detectable at exposure levels resulting in maternal hair mercury levels below 10 ppm (Grandjean et al., 1997). Although only limited dose response data are available from this study, the median mercury maternal hair concentration for the cohort was 4.3 ppm and control group formations were made with mothers who had lower exposures and mercury hair concentrations below 3.0 ppm (Grandjean et al., 1997, 1998).

Based on available data, a tolerable daily intake for the populations of greatest concern (women of child bearing age and their infants) could be determined using mercury maternal hair or mercury cord blood exposure data. The pharmacokinetic variability would

be less if mercury cord blood levels from the Faroe Islands cohort were used since empirical cord blood data would not require using mercury hair levels as the exposure metric to estimate maternal blood mercury, which is itself an estimate of cord blood. Also, in a compartment model that relates exposure to target organ dose, mercury cord blood; if strongly associated with mercury maternal blood, would be just one compartment removed from the target organ. At present, for the Faroe Islands study, only very limited dose response data and some data on the distribution of maternal hair mercury levels are available, while data on the relationship between maternal and cord blood levels for mercury are not yet available. Only the geometric average cord blood level of 22.9 µg/l has been provided, without any variance data.

Previous works have suggested that the average mercury cord blood levels are 20 – 30% higher than mercury maternal blood levels (Kuhnert et al., 1981). Dennis and Fehr (1975) analyzed paired maternal and cord blood samples for mercury from fish consuming women in northern Saskatchewan (n = 43) and non-fish consuming women living in southern Saskatchewan (n = 45). There was a positive association between mercury maternal and mercury cord blood levels in both regions with the correlation coefficients being 0.45 and 0.87 for the south and north, respectively. Only in the north though, was the mean mercury level significantly different ( $p < 0.01$ ) between maternal and cord blood samples. The cord blood samples were higher for the north sample group with the slope of the regression being 1.3. Kuhnert and associates (1981) re-addressed this issue of maternal and cord blood mercury level differences using gas chromatography techniques in a small group study (n = 29). Methylmercury levels in both plasma and erythrocytes were investigated with 30% more methylmercury observed in fetal erythrocytes than in maternal erythrocytes, while



plasma levels were not significantly different. “Total” mercury concentrations in blood were calculated and compared with other studies. Results indicated that “total” mercury levels in fetal cord blood are 13% to 24% higher than those in maternal blood. Kuhnert and associates (1981) also suggested that fetal cord whole blood contained 32% more methylmercury than maternal whole blood which is similar to the increase observed between fetal and maternal red blood cells. Although the sample size is a limitation of this study, this work does suggest that the ratio of mercury cord blood levels to mercury maternal blood levels is greater than one.

Further indication that the ratio may not be equal to one can be found in data presented by Cernichiari and co-workers (1995) indicating that mercury infant blood levels are on average twice that of maternal blood. These data, however, may only be used to suggest that differences in mercury blood levels may exist since the mercury levels in fetal blood and infant blood can not be directly compared because they are not identical; for example, fetal hemoglobin, manufactured in the red blood cells of the fetus and infant compose 50% to 90% of the hemoglobin in the newborn, however, it is mostly replaced by adult types (A<sub>1</sub> and A<sub>2</sub>) by age six months (Fischbach, 1996).

By applying the ratio of mercury cord blood levels to mercury maternal blood levels suggested by data sets described above (1.3) to the available cord blood value of 22.9 µg/l, a maternal blood value of 17.6 µg/l is obtained. A daily intake of 0.36 µg/kg/day is then derived using the following algorithm relating mercury levels in maternal blood to a daily intake level (WHO, 1990; ATSDR, 1997):

$$C = \frac{A_D * A_B * d * W}{b * V}$$

where,

C = mercury concentration in blood (mg/l),

A<sub>D</sub> = percent of mercury intake in diet that is absorbed (95%),

A<sub>B</sub> = percent of the absorbed amount that enters the blood (5%),

d = daily dietary intake (mg/kg),

b = elimination constant (0.014),

V = volume of blood in 60 kg woman (4.2 l), and

W = average body weight for women (60 kg).

Studies that have compared mercury maternal hair levels with mercury blood levels have produced various ratios ranging from 140 to 415 (Berglund et al., 1971; Birke et al., 1972; Davidson et al., 1995; Den Tonkelaar et al., 1974; Kershaw et al., 1980; Sherlock et al., 1982). Within this range of ratios (which differ by approximately three), the most frequently used value has been 250. Blood samples were not obtained from the New Zealand cohort; however by applying this most frequently cited hair to blood ratio along with the 6 ppm maternal hair mercury value, a daily intake of 0.5 µg/kg/day is obtained by using the algorithm described by WHO (1990) and ATSDR (1997). Also, by applying the hair to blood ratio of 250 to the Faroe Islands maternal hair levels of 4.3 and 10 ppm, a daily intake range of 0.35 to 0.8 µg/kg/day is obtained. The geometric average maternal hair level of 4.3 ppm was used because the regression relationship between methylmercury exposure and adverse effects was derived from the entire cohort and the average value reflects that cohort

(notwithstanding that the regression may be driven by values above or below the average value), while 10 ppm represents the cutoff value used in the bivariate categorical analyses which showed a significant difference for methylmercury effect above and below that value. Although only limited dose response data are available for the Faroe Islands population, the average maternal hair mercury level and the maternal hair mercury level of 10 ppm, below which early dysfunction is detectable, may provide a range that encompasses the intake level considered tolerable for that population. Also, this range encompasses the daily intake level of 0.5 µg/kg/day obtained using cord blood data and intake rate obtained from the New Zealand cohort results.

The use of these data to derive a tolerable daily intake level, in light of the Seychelles Islands study results, raises the issue of why seemingly similar exposure levels can result in different outcomes. Confounders or other subtle factors presently not properly delineated may be responsible for differences seen between populations that have similar mercury-hair concentrations and that are exposed in similar manners. Along with possible population differences, some of the observed discrepancies may be due to the exposure metric used in the different studies, either cord blood or maternal hair. Also, these exposure levels may be at or near the effect level such that other factors may determine if mercury exposure at these levels impacts brain function. Both the Faroe Islands and Secychelles Islands studies removed individuals with severe deficits from its cohorts, based on very specific conditions, prior to initial analysis of the data. And though outliers were treated differently between the two studies, both the Faroe Islands and Seychelles Islands studies can be considered to be relatively inclusive in addressing the range of variability in dose-response which may be expected in populations such as those in the US. Both studies were specific to the most

sensitive portion of the population (mother-infant pairs) and included those with relatively moderate rates of fish/whale consumption as well as those who would be considered to consume elevated quantities in the US. One limitation in interpretation of these studies, however, is that both implicitly assume that adverse effects from methylmercury are associated with average daily intake. Both studies also assume that mercury-hair concentrations and, to some extent, mercury-blood concentrations reflect average intake. Though this assumption may be correct in that these metrics do reflect average intake, they may not reflect intake over the same time period (i.e. temporal differences in hair and blood measurements). It is possible that the adverse effects of methylmercury exposure are more directly related to the magnitude of peak exposure, as could result from one or a few closely spaced meals of fish with high mercury concentration, rather than to average exposure level. If peak exposure were an important determinant in predicting adverse effects of methylmercury exposure, then exposure calculations based on average exposure metrics could result in significant exposure misclassification. This, in turn, would reduce the predictive power of these studies. Unfortunately, none of these three primary studies collected consumption frequency information by fish species, which could have been useful in distinguishing the influence of average versus peak exposure.

Considering the totality of evidence from these studies with respect to effects observed at less than 10 ppm maternal hair mercury levels and the populations studied, determining the exact value that should be applied to address various uncertainties is problematic. The sensitive endpoints are impaired neurological development and long-term and/or delayed sequelae. The studies used to derive a tolerable daily intake best address the former with long-term and/or delayed sequelae being effects that have only been observed

and/or studied in primates and in the catastrophic exposure to communities in Japan (Rice, 1992, 1996; Harada, 1995). There is uncertainty associated with toxicodynamic variations across the populations, although given the cohort sizes and types, this variation may be small (Renwick, G., 1993; Dourson et al., 1996; ATSDR, 1997). The inter-individual pharmacokinetic variability associated with determining a tolerable intake level based on hair mercury levels could be accounted for through the use of an uncertainty factor of three applied to a central tendency estimate of the intake does corresponding to the maternal hair concentration (Renwick, G., 1993; Stern, 1997; Clewel et al., 1999). In total, these variabilities and the lack of ability to address long-term and/or delayed sequelae warrant an additional reduction of one order of magnitude. The various elements associated with this uncertainty will require re-evaluation as further data are made available. Although this value may be too large, a present justification for a smaller value can not be garnered from available scientific evidence. As a result, the tolerable daily intake becomes the range of values from 0.035 to 0.08 µg/kg/day.

#### **Exposure analysis for recreational anglers (freshwater fish species)**

##### **Angler Survey Results:**

Surveyed anglers were primarily members of two-adult households (84%). Of all households, 29% had children under eighteen years of age. In 96% of two adult households, both adults consumed Lake Roosevelt caught fish. Sixty percent of anglers interviewed were considered by the interviewer to be over fifty years of age. Most anglers were male (90%) and Caucasian (97%). Only 2.4% of respondents were identified as Native American. It is possible that Native Americans fishing the lake were not surveyed since chosen survey

locations were primarily off tribal lands. However, only a limited portion of the Spokane Tribe of Indians and the Colville Indian Nation use the lake as a fisheries resource.<sup>1</sup>

Yearly meal frequencies for kokanee(*Oncorhynchus nerka*), rainbow trout (*Oncorhynchus mykiss*), walleye (*Stizostedion vitreum*) and bass (*Micropterus dolomieu*) were derived for each angler (n=348). Distribution data indicate that for individual fish species, approximately 90% of respondents consume 39 or fewer meals per year (Table 1). The total number of meals for all fish species consumed over the course of a year for each angler (n=348) was also determined (Table 2). The data indicate that more than 90% of respondents consume 103.2 meals (2 meals/week) or less per year, and that nearly 75% of respondents consume 48 or fewer meals per year.

Fillets were the primary portion consumed, as indicated by 84% of respondents (n=377). Thirty percent of respondents consumed whole-gutted fish. Few anglers consumed fish skin, eggs, or fish head, while no one indicated eating fish bones or guts. Respondents (n=377) also indicated that primary cooking methods were pan frying (77%), smoking (41%), and baking (39%). Few anglers prepared fish by broiling, roasting, barbecuing or steaming, and no anglers consumed raw fish or fish in soup.

In an effort to gain insight into amount consumed per meal, 1994 surveys contained a general query about the number of fillets consumed per meal. A similar question was asked in 1995 surveys, although the query asked about consumption on a per species basis and on the number of fish, not fillets, consumed per meal. Results from 1994 indicate that greater than 95% of respondents (n=176) consume one or two fillets per meal (Table 3). In 1995, one or fewer rainbow trout was eaten per meal by approximately 80% of respondents, and one or

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<sup>1</sup> Personal communications; K. Underwood, Spokane Tribe, P.O. Box 100, Wellpinit, WA, 1996 and B. Aripa, Confederated Tribes of Colville, P.O. Box 150, Nespelem, WA, 1996.

fewer walleye was consumed per meal by 70% of respondents. Approximately half of the respondents who consume kokanee and bass consumed one fish during a meal while the remaining half consumed two fish (Table 3).

#### Mercury Fish Tissue Results:

The principal sport-fish species in Lake Roosevelt are kokanee, rainbow trout and walleye (McDowell and Griffith, 1993). Walleye have been reported to contain the highest concentrations of mercury among sport-fish species routinely caught in Lake Roosevelt (Johnson et al., 1988; Serdar et al., 1993; Munn et al., 1995). In a study conducted in 1994 by Munn et al. (1995), mercury concentrations in skinned walleye fillet composite samples (n = 34) ranged from 0.11 to 0.44 mg/kg (wet weight), with an overall reported mean mercury concentration of 0.34 mg/kg (SD=0.07) (Munn and Short, 1997). Results of subsequent sampling in 1998 suggest that mercury levels in walleye declined by 59 percent to a catch weighted average concentration of 0.17 mg/kg (wet weight).<sup>2</sup> Based on a walleye/rainbow trout mercury concentration ratio of 0.21, rainbow trout were estimated to contain 0.04 mg/kg mercury (Norecol, 1989). The mean mercury concentration in kokanee was assumed to be the same as that for rainbow trout due to similarities in life histories between species (Wydowski and Whitney, 1979). The mean mercury concentration in bass from the analysis of five composite samples consisting of five fish each was 0.28 mg/kg (wet weight) (SD = 0.19) (Munn et al., 1995).

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<sup>2</sup> Personal communication, M. Munn, United States Geological Survey, Water Resources Division, Tacoma, WA, 1999.

### Individual Mercury Exposure Results:

To estimate population exposure to mercury, the proportion of the angling population potentially exposed to mercury levels above the TDI was estimated using data from the sample of anglers surveyed during 1994 and 1995. For each angler surveyed who ate at least one species of fish ( $n = 343$ ; five individual records of the original 348 lacked complete fish species consumption information), a daily intake of mercury was estimated for each species of fish caught. The daily intake of mercury for walleye, kokanee, rainbow trout and small mouth bass, was calculated as the product of the number of meals per month when that species was consumed, the usual number of fillets consumed at a meal, the average weight of a fillet of that species, and the average fish tissue mercury concentration for that species in Lake Roosevelt. The total daily average mercury intake was estimated for each person by summing the estimated intakes due to each of the four fish species. This value was converted to units of micrograms per kg body weight per day (assuming an average adult body weight of 70 Kg) and compared to the upper bound of the TDI ( $0.08 \mu\text{g/kg/day}$ ).

*Fish consumption.* During 1994, surveyed anglers averaged 1.7 fillets per meal (Table 3). In 1995 the average number of fillets consumed per meal across species was 2.6, assuming two fillets per fish. As determining actual fillet consumption amounts per meal was not a primary focus of this study (fish fillet/steak models were not used) a protective approach was used to determine fish consumption rates. For anglers surveyed in 1995, the species-specific values, along with the conversion factor of two fillets per fish, were used for the consumption estimates. For the anglers surveyed in 1994, the average consumption rate from the 1995 angler survey of 2.6 fillets per meal was used instead of the lower average value of 1.7 fillets per meal.



*Fillet weights.* The average mass of fillets consumed by anglers was estimated using species-specific fillet:whole fish weight ratios and angler-caught whole fish weights. An average fillet weight to whole fish weight ratio was calculated for bass (0.0935), rainbow trout (0.1295), and walleye (0.1418) using data collected by Munn et al. (1995). The fillet-to-whole fish weight ratio calculated for rainbow trout was also used for kokanee because of similar morphology between species and due to lack of kokanee data. For walleye, the mean fillet-to-whole fish ratio was weighted by the number of samples within each size class. Mean fillet weights for angler-caught small mouth bass, Kokanee, rainbow trout, and walleye in 1995 are estimated to be 58.46 (SD=48.57), 157.42 (SD=46.91), 122.80 (SD=59.18), and 55.45 (SD=36.12) grams, respectively.

*Data analysis.* SAS (SAS Corporation, Cary, NC) was used to generate the mercury intakes for the population of anglers and to develop summary statistics of the mercury intake and the ratio of individual intakes to the TDI.

Analysis of the consumption data on an individual basis indicated that nearly half the fish consumed were rainbow trout (49%) as compared to 28% for walleye. While walleye are responsible for approximately 40% of the mercury intake, rainbow trout provide 36% of total mercury uptake due to it being frequently consumed. Of the 343 individual anglers surveyed, nearly 87% (298 individuals) had mercury intake levels at or below the upper bound of the TDI. Only 21 individuals had mercury intake rates between 0.06 and 0.08  $\mu\text{g/kg/day}$ . Ten individuals had intake levels between two and four fold higher than the TDI, while the remaining individuals ( $n = 35$ ) had daily mercury intake due to fish consumption between the upper bound of the TDI and twice the upper bound. This portion of the population ate specific species at a rate three to five fold higher than the mean yearly

consumption rate for the population surveyed. Also, these individuals consumed fish in greater quantity during a meal; depending on the fish species, 60% to 90% of the individuals consumed four fillets per meal, which is considerably higher than the mean for anglers surveyed in 1995 (2.6 fillets/meal). Nearly all of the individuals exceeding the TDI were adult males estimated to be greater than fifty years of age.

### **Exposure analysis for recreational shore and boat anglers (saltwater fish species)**

Landolt and co-workers (1985, 1987) determined mercury concentrations in various fish species in connection with fish consumption data. The 1985 shore angler data were compiled so that populations with the highest consumption rates for a particular fish species were categorized and mercury intake levels for each population were determined (Table 4). Landolt and co-workers also determined which species were most often consumed from specific locations. Thus consumption values were added; for Pacific hake, starry flounder and Pacific cod since consumption rates for these species were higher at a specific location than rates determined for particular ethnic populations (Table 4). Boat angler data were obtained in three locations and intake levels for individuals consuming fish from these locations were determined (Landolt et al., 1987) (Table 5). Although these data do not provide consumption rates by ethnicity but by location, ethnic origin data for two locations indicate that 86% of the population was Caucasian (Landolt et al., 1987). Consumption rates for both studies were expressed as geometric mean grams of cleaned fish available for consumption per person per day during the time period each species was present in fishery (Tables 4, 5). Total mercury intake levels for all fish species among both shore and boat anglers were less than the TDI, even with the assumption that the fish were consumed throughout the year.

More data on chemical contaminants in Puget Sound fish have become available recently (West and O'Neill, 1995; PSWQA, 1995; PSWQAT, 1998; O'Neill et al., 1998). These data consist mainly of composite samples for Pacific salmon, English sole and rockfish obtained from over fifty locations during the 1990's. Multiple linear regression analysis indicated that age, not location, is the primary variable associated with mercury concentrations in English sole and Pacific salmon (West and O'Neill, 1995). For example, age of English sole accounts for approximately 70% of the variability of mercury concentrations in muscle tissue, with sediment concentrations of mercury accounting for 4% (West and O'Neill, 1995; PSWQA, 1995; PSWQAT, 1998; O'Neill et al., 1998). Moreover, chinook salmon have the highest mercury concentrations of the two salmon species tested since they remain in a saltwater environment (e.g., open ocean) longer than coho salmon (typically 4 years or longer) (Wydoski and Whitney, 1979). For rockfish, age and location were important variables, whereas it is unclear as to the effect of source concentration (mercury in sediment) (West and O'Neill, 1995; PSWQA, 1995; PSWQAT, 1998; O'Neill et al., 1998). As a result, the authors were unable to indicate whether the rockfish with high mercury concentrations (age corrected) only come from contaminated locations (industrialized), although it appears that most do. One location (industrialized) was sampled for fish mercury concentrations by O'Neill and co-workers for which the Landolt study had geometric mean consumption rates. The mercury fish tissue level for coho and chinook was based on the median value of eighteen composite means having five salmon per composite; levels were 0.04 mg Hg/kg and 0.10 mg Hg/kg for coho and chinook salmon, respectively. Mercury intake values were calculated to be 0.02 µg/kg/day and 0.09 µg/kg/day for coho and

chinook salmon, respectively. Thus, the intake value for chinook salmon slightly exceeds the TDI, while the intake value for coho does not.

### **Exposure analysis for Tulalip and Squaxin Island Tribes**

Consumption data for the Tulalip and Squaxin Island Tribes by fish category (anadromous, pelagic, bottom) have also been collected (Toy et al., 1996). These three fish categories represent nearly all of the fish types consumed by both tribes as other fish such as trout, manta ray, shark and canned tuna are consumed infrequently and/or by few individuals. The 90<sup>th</sup> percentile consumption rates from consumption distributions for each of the three categories of fish consumed were combined with fish contamination data from O'Neill and co-workers to determine level of exposure (Table 6). Even though some fish were obtained from grocery stores and restaurants, all fish were assumed to be angler caught. Toy and co-workers stratified total finfish consumption rates by age and sex with no statistical differences observed at median consumption rates. The 90<sup>th</sup> percentile values were combined with median mercury fish concentrations to derive intake levels.

For both the Tulalip Tribes and the Squaxin Island tribe, intake values for anadromous fish (defined only by the consumption of coho or chinook salmon), are at or above the TDI. The consumption values obtained by combining the values from each fish category are 1.696 g/kg/day for the Tulalip Tribes and 1.921 g/kg/day for the Squaxin Island tribe, respectively. These values correspond to the 90<sup>th</sup> percentile total finfish consumption values of 1.779 g/kg/day and 1.828 g/kg/day for the Tulalip Tribes and Squaxin Island tribe population, respectively (Toy et al., 1996). The corresponding methylmercury intake values, depending on fish species consumed, range from 0.11 – 0.2 µg Hg/kg/day for the Tulalip

Tribes and 0.11 – 0.22 µg Hg/kg/day for the Squaxin Island tribe (Table 6). Salmonid consumption is the primary cause for intake values exceeding the TDI range as the upper bound of the TDI (0.08 µg Hg/kg/day) is exceeded by 8 – 14% of the Tulalip Tribe population when consuming salmon only and 10 – 25% of the Squaxin Island tribe population when consuming salmon only (Table 6).

While these values represent one result, it must be noted that the study by Toy and co-workers required a sample size of 150 for the Tulalip Tribes and 120 for the Squaxin Island tribe to achieve sample sizes that would provide reasonable precision of estimates of mean consumption (upper and lower bounds of confidence intervals lying within 20% of an estimated mean). The sample sizes achieved were 73 and 117 for the Tulalip Tribes and Squaxin Island tribe, respectively. Thus, the distribution for the Tulalip Tribes may misrepresent the total population distribution. This is noteworthy in that only four individuals separate the 86<sup>th</sup> from the 90<sup>th</sup> percentile consumption rate values for anadromous fish, with a rise in consumption rate values from 0.63 to 1.429 g/kg/day for the 86<sup>th</sup> and 90<sup>th</sup> percentiles. If the 86<sup>th</sup> percentile value 0.63 g/kg/day is used, the estimated mercury intake level ranges from 0.07 – 0.12 µg Hg/kg/day for the Tulalip Tribes (as compared to 0.11 – 0.20 µg Hg/kg/day using the 90<sup>th</sup> percentile).

### **Exposure analysis for Suquamish Tribe**

Consumption data have recently been collected for the Suquamish Tribe of the Port Madison Indian Reservation (The Suquamish Tribe) in which consumption patterns were determined for various pelagic and salmonid fish species (The Suquamish Tribe, 1999). This study required a sample size of 158 to provide reasonable precision of estimates of mean consumption. After multiple attempts to obtain survey information from possible

respondents, 92 surveys were completed. Inherently, this data set has similar difficulties to those observed with the Tulalip Tribes data. It is not known if 92 individual respondents (58% of required sample size) are a representative sample; especially since demographic data on non-respondents is unavailable.

The 90<sup>th</sup> percentile adult consumption rate for consumers of salmon species and for the fish group containing halibut, rockfish and sole are 1.680 and 0.392 g/kg/day, respectively (Table 6). Unlike the other two Native American groups where median tuna consumption was extremely low, tuna (fresh/canned) is consumed by this population, with the 90<sup>th</sup> percentile consumption rate being 0.346 g/kg/day. The summation of these 90<sup>th</sup> percentile totals is 2.418 g/kg/day which corresponds to the 90<sup>th</sup> percentile total for all finfish which is 2.526 g/kg/day. Although some fish were obtained from grocery stores and restaurants, all fish were assumed to be angler caught.

Individual fish species consumption data from the respondents was also collected in this study. These data indicate that chinook salmon is not eaten in greater quantities than other salmonids. In the finfish category (consisting of red snappers, rock fish, sole and halibut), only ten respondents indicated that they consumed rock fish, while 3, 20 and 74 individuals consumed red snappers, sole and halibut, respectively. The halibut caught by the Suquamish tribe should have background levels of mercury as they are obtained from open waters (Strait of Jaun de Fuca) in spring/early summer when halibut are searching for food and before the halibut migrate back north into open ocean in mid-summer.<sup>3</sup> As rock fish are consumed by few respondents and halibut contaminant data are unavailable, the English sole contaminant level derived by O'Neill and co-workers was used in the exposure analysis for this population (Table 6). Additional contaminant data from O'Neill and co-workers for

coho and chinook salmon along with a mercury tuna level of 0.17 mg/kg provides for mercury intake levels that range from 0.16 to 0.25 µg Hg/kg/day (Table 6)(Yess, 1992). While the percent of the population exceeding the TDI could not be determined, 75<sup>th</sup> percentile consumption rates yielded an intake range of 0.08 to 0.13 µg Hg/kg/day. The exposure values based on 90<sup>th</sup> percentile consumption rates for individuals consuming salmon, either coho or chinook, are at or above the TDI (Table 6). For those ten individuals consuming rockfish, their exposure is also at or above the TDI, depending on the type of rockfish consumed (Table 6).

## **Discussion**

The goal of this study was to determine if various populations in this state are consuming contaminated fish in quantities that could possibly result in deleterious outcomes. To accomplish this, consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species were examined. The consumption rates were used with fish contamination data to analyze exposure to methylmercury. Concomitantly, a TDI was derived for methylmercury (0.035 to 0.08 µg/kg/day). The TDI is unlikely to result in adverse health effects and is based on scientific studies investigating sensitive endpoints in children of mothers who consume fish over prolonged periods of time. The results from the exposure analyses for the diverse populations were then compared with the TDI to determine if certain populations were exceeding this health effects value.

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<sup>3</sup> Personal communication , J. Zischke, Suquamish Tribe, P.O. Box 498, Suquamish, WA, 2000.

### **Exposure to recreational anglers (freshwater fish species)**

Angler consumption patterns were determined and used with mercury fish tissue contamination data to determine if this Lake Roosevelt fish consuming population is exposed to levels of mercury that are above levels deemed to not result in adverse health effects. Results from the exposure analysis indicate that, at a minimum, 87% of the anglers surveyed have intake levels below the upper bound of the TDI. This percentage value was not sensitive to changes in the number of fillets consumed per meal as there were few individuals consuming quantities of fish resulting in mercury intake levels just below or above the upper bound of the TDI (0.08 µg/kg/day). Also, without a significant change in mercury fish tissue concentrations, the percentage of individuals consuming fish resulting in mercury intake levels just below or above the upper bound of the TDI will not change dramatically. Those individuals exceeding the TDI were those who ate fish more frequently and in greater quantity than the average angler. Due to the sampling method used, a disproportionate number of individuals who fish more frequently may have been interviewed. Nearly all individuals exceeding the TDI were adult males estimated to be greater than fifty years of age. If the spouses of these older adult males are of approximately the same age, they also would not represent a population of concern (women of child bearing age). Further distribution data on female age and consumption patterns would provide additional support for this conclusion. As rainbow trout already represent nearly half of the fish consumed by these anglers, recommending that trout be consumed in even greater quantity (since they are less contaminated than walleye), may not result in a significant reduction in individual mercury intake. Based on the age structure of the individuals interviewed, there does not appear to be a population consisting of women of child bearing age consuming fish resulting



in intake levels above the TDI. As the benefits of consuming fish as a source of protein compared to other protein sources is well documented, suggesting that older adults, male or female, consume less fish may have a deleterious impact. However, to reduce exposure to mercury in these individuals that eat fish more frequently and in greater quantity, educational efforts informing them to consume a variety of fish species from a variety of locations could be of benefit.

### **Exposure to recreational shore and boat anglers (saltwater fish species)**

The exposure analyses for the recreational shore and boat anglers suggests that mercury intake levels are below the TDI. However, anglers consuming chinook salmon from one particular location (industrialized) have methylmercury exposures ( $0.09 \mu\text{g/kg/day}$ ) just slightly exceeding the TDI. This result is significant for two reasons. First, concentrations of methylmercury are considered to be uniform for this species in Puget Sound suggesting that anglers representing our population of concern who consume greater than  $0.8 \text{ g/kg/day}$  of chinook salmon would be exceeding the TDI. Second, the result of this exposure analysis was based on a geometric mean value, with no distribution data being available, indicating that all individuals consuming above this value are also exceeding the TDI. Further consumption pattern data will be required to determine if this historical consumption data is still valid and to determine if recreational anglers who represent our population of concern are exposed to mercury levels above the TDI. Presently, determining a clear course of action is difficult. Evidence does not exist to warrant a recommendation that recreational anglers who frequently consume chinook salmon should reduce their consumption rates, however, this is an issue definitely requiring attention.

### **Exposure to Tulalip and Squaxin Island Tribes**

Several conclusions can be derived from the Tulalip Tribes and Squaxin Island tribe data. The 90<sup>th</sup> percentile total finfish consumption values for both tribal groups are approximately equal to the sum of 90<sup>th</sup> percentile values for each of the fish categories, suggesting that individuals consuming elevated amounts of fish from one category may be eating elevated quantities from the other two. Although the contribution of mercury intake from pelagic and bottom fish may be small compared to anadromous fish, the results suggest that at least 8 – 14% and 10 – 25% of the Tulalip and Squaxin Island tribal members, respectively, have mercury intake levels above the TDI. Clearly, individuals within both populations are exceeding the TDI, however, the percent population value of 8 – 14% for the Tulalip Tribes that may have intake levels above the TDI will require validation through consumption distribution data. Further narrowing the range for either population is difficult as the salmonid consumption distribution (i.e. chum, steelhead, sockeye, pink, coho and chinook) was not collected and fish tissue mercury data are only available for coho and chinook salmon.

Given the cultural, spiritual and historical significance of fish consumption by tribal members, the suggestion of dietary changes may produce no clear benefit and could even result in deleterious health effects. Recommending changes must be weighed against the benefit of cultural events such as tribal ceremonies which are attended frequently and are a significant and important sources of fish. Present consumption data indicate that the mercury intake range in Tulalip Tribes (using the 86<sup>th</sup> and 90<sup>th</sup> percentiles) is 0.07 – 0.2 µg/kg/day, with the latter value based on the assumption that all individuals consume only chinook salmon. Until improved consumption data are available to better determine the percent of the population exceeding the TDI, most if not all individuals within this population could

achieve mercury intake levels at or below to the TDI while maintaining their cultural heritage by consuming other anadromous fish as alternatives to chinook.

The data for the Squaxin Island tribe present a much different dilemma. Results from the exposure analysis indicates that many individuals (25% of the Squaxin Island tribe) are consuming anadromous fish in quantities that may result in a mercury intake above the TDI. This conclusion is based on the assumption that chinook salmon are the only anadromous fish consumed. The consumption of other salmonids (i.e. chum, coho, steelhead, sockeye and pink) could reduce this value to approximately 10% of the population. Using the 90<sup>th</sup> percentile values also reflects that individuals have intake levels above the TDI. At a minimum, women of child bearing age within this population should be encouraged through educational efforts to consume salmonids other than chinook. As changing consumption of salmonid species will still result in intake levels above the TDI, the potential for adverse outcome remains. Simply suggesting that fish be consumed in lesser quantities is not, however, necessarily a prudent public health recommendation. Recommending a change in diet away from nutritional foods such as fish does not imply that the replacement will be equally beneficial. Also, as stated, cultural, spiritual, and historical practices must be considered. Educational efforts could be provided to encourage the broadening of fish species consumed to include freshwater fish species or pelagic fish that possibly have lower mercury concentrations (e.g. cod, pollock, herring, perch). Also, as hair analysis is presently the exposure metric most frequently relied upon, hair levels of methylmercury in the women of childbearing age who consume fish in excess of 1.0 g/kg/day should be monitored. These data could be used along with educational efforts as excellent tools for properly protecting the health of this population.

## **Exposure to the Suquamish Tribe**

The Suquamish tribe data suggest that individuals within the population have mercury intakes above the TDI. As with the above tribes, the 90<sup>th</sup> percentile total finfish consumption values are approximately equal to the sum of 90<sup>th</sup> percentile values for each of the fish categories suggesting that individuals consuming elevated amounts of fish from one category may be eating elevated quantities from the other two. The mercury intake range for the 90<sup>th</sup> percentile of these 92 individuals was estimated to be (0.16 to 0.34 µg/kg/day) with approximately 25% of those interviewed having mercury intake levels above the TDI. Two factors suggest that the actual mercury intake levels may be nearer the lower end of this range. First, there are four types of salmon species known to be regularly consumed by this population. The coho salmon mercury tissue concentration (0.05 mg Hg/ kg fish) may better represent an appropriate mercury fish tissue concentration with which to determine mercury intake levels than that of chinook salmon, which is considered to have the highest levels of mercury (1.0 mg Hg/ kg fish)(Table 8). Second, greater than 50% of the fish consumed from the category containing halibut, sole, rockfish, flounder, and red snapper came from groceries and restaurants. Even considering the lower end of the range established from the exposure analysis (0.16 µg/kg/day), the TDI is still being exceeded by two-fold. When using this reasoning with the 75<sup>th</sup> percentile consumption rates, the mercury intake level becomes 0.08 µg/kg/day, which is equivalent to the upper end value of the TDI. Thus, ten percent of the surveyed population sampled exceed the TDI by two-fold while 25% are at or above the TDI.

Inherently, this data set has similar difficulties to those observed with the Tulalip Tribes data as it is not known if 92 individual respondents are a representative sample. However, until improved consumption distribution data become available, women of child

bearing age within this population should be encouraged through educational efforts to consume salmonids other than chinook. Many individuals having intake levels above the TDI could reduce mercury intake levels, while maintaining their cultural heritage, by minimizing chinook salmon consumption. Rockfish consumption needs to be decreased by those consuming elevated quantities in this population. Eight of the ten individuals consuming rockfish, consume at the rate of approximately 0.124 g/kg/day (75<sup>th</sup> percentile) or less. Assuming Quilback and Copper rockfish are eaten equally, this results in a mercury intake level of 0.03 µg/kg/day or less, which is well below the TDI. However, those two individuals responsible for the 90<sup>th</sup> percentile consumption value of 0.789 g/kg/day have mercury intake levels of 0.18 µg/kg/day. As the rockfish contaminant level data did not come from the specific fishing areas used by this population, rockfish that this population regularly consume should be analyzed for mercury so that an accurate determination can be made of how much is being consumed. Many of the same recommendations made with respect to the other tribes applicable here, including consumption of other fish species having lower mercury concentrations and monitoring hair levels of methylmercury in the women of childbearing age who consume fish in excess of 1.0 g/kg/day as a preventive health measure.

## **Conclusion**

We examined consumption rates of three Native American populations and two recreational fishing populations consuming freshwater or saltwater fish species. The consumption rates were used with fish contamination data to analyze exposure to methylmercury and to determine if any of these populations exceeded a derived TDI for methylmercury (0.035 to 0.08 µg/kg/day). Results from the exposure analyses indicate that

recreational anglers consuming freshwater species have exposure levels below the TDI as do recreational anglers consuming saltwater fish species with the exception of those consuming chinook salmon from one particular location (industrialized). The exposure analyses also indicate that many within the Native American populations exceed the TDI and that this occurs even though the mercury concentrations in certain fish species are comparable to concentrations found in fish from open waters where “background” levels are assumed. As with any TDI, changes to the value must be made as further data become available. Without maintaining diligence in this area, the result could be public health protection through a TDI that is too high which allows for individuals to be exposed to deleterious levels or through a TDI that is too low which would provide for a large gap between acceptable intake levels and those levels that cause toxic effects. With respect to mercury, it is imperative that this gap be minimized as a TDI that is too low will be a public health detriment as it results in recommendations that restrict or alter the consumption patterns of healthy food sources.

Also of import are “background” mercury levels present in salmon resulting in populations being exposed to mercury levels above the TDI. Regardless of whether the levels are “background” or above, public health protection can only be achieved by ensuring that exposure levels remain within present protective and accepted values, even if this impacts the consumption patterns of a particular fish type (chinook salmon) which is otherwise considered a very healthy food source. This is however not to suggest that other protein sources beside fish should be considered, but only that a variety of fish be consumed as the benefits of this protein source compared to others is well documented. In the case of Washington state, we must ensure that various salmon species are available for consumption so that chinook salmon are consumed by choice and not necessity.

The approaches used to protect individuals require difficult decisions when cultural and historical considerations must be considered or when intake levels just slightly exceed a value or set of values deemed to be protective. In this study, recommendations for how best to serve the public health needs of these populations regarding methylmercury exposure have been provided. The study outcomes presented herein allow state health departments and local health jurisdictions to develop intervention and education strategies to protect individuals, especially women of child-bearing age, from overexposure to methylmercury.

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**Table 1.** Species-specific fish meals consumed per year for individuals (n = 348) consuming that particular species, Lake Roosevelt, WA, 1994-95.

Meals/Year	Kokanee (n=140)	Rainbow Trout (n=299)	Walleye (n=231)	Bass (n=99)
>0 - 6	27 (19%)	109 (36%)	57 (25%)	18 (18%)
8 - 14	31 (22%)	63 (21%)	52 (23%)	22 (22%)
15 - 20	23 (16%)	28 (9%)	33 (14%)	22 (22%)
22 - 28	22 (16%)	46 (15%)	44 (19%)	16 (16%)
30 - 39	20 (14%)	28 (9%)	26 (11%)	13 (13%)
42 - 52	14 (10%)	19 (6%)	15 (6%)	7 (7%)
>52	3 (2%)	6 (2%)	4 (2%)	1 (1%)
mean $\pm$ s.d.	22 $\pm$ 18	18 $\pm$ 19	19 $\pm$ 16	20 $\pm$ 14

**Table 2.** Total number of fish meals consumed per year by anglers, Lake Roosevelt, WA, 1994-95.

Consumption Frequency (meals/year)	≤12	>12-24	>24-48	>48-103.2*	>103.2	mean ± s.d.
Respondents (n=348)	121 (35%)	65 (19%)	67 (19%)	68 (20%)	27 (8%)	42.2 ± 47.7

\*Reflects two fish meals per week based on 4.3 weeks per month.

**Table 3.** Fillets consumed per meal (1994 data) and fish consumed per meal on a per-species basis (1995 data), Lake Roosevelt, WA, 1994-95.

Fillet or Fish /Meal	1994 Data	1995 Data			
	Fillets n=176	Kokanee n=48	Rainbow Trout n=124	Walleye n=108	Bass n=21
≤ 0.5	0 (0.0%)	1 (2%)	26 (21%)	2 (2%)	0 (0%)
1	59 (33.5%)	21 (44%)	72 (58%)	74 (68%)	10 (48%)
2	110 (62.5%)	26 (54%)	26 (21%)	28 (26%)	11 (52%)
>2	7 (4.0%)	0 (0%)	0 (0%)	4 (4%)	0 (0%)
mean ± s.d.	1.7 ± 0.6	1.5 ± 0.5	1.1 ± 0.5	1.3 ± 0.6	1.5 ± 0.5

**Table 4.** Puget Sound Shore Angler Data (Landolt et al., 1985). Estimated mercury intake levels for populations consuming highest levels of particular species.

Fish	Population	Months fish predominantly consumed	Consumption rate (g/kg/day)**	Fish tissue Hg concentration (mg Hg/kg fish)	Estimated Hg intake** (µg/kg/day)
Squid	Chinese-Japanese	-	1.0	0.10	0.01
Tomcod	S.E. Asian	Aug – Mar	0.3	0.005	0.002
Walleye-Pollock	S.E. Asian	Oct – Feb	0.38	0.016	0.006
Pacific hake	U.S. (Cauc.) Elliot Bay*	Jul – Nov	0.35	0.005	0.002
			0.57	0.005	0.003
Sablefish	U.S. (Black)	-	0.78	0.013	0.01
Starry Flounder	U.S. (Cauc.) Edmonds*	Apr – June	0.33	0.02	0.007
			0.83	0.02	0.017
Pacific Cod	U.S. (Cauc.) Sinclair Inlet*	Jan – Dec	0.48	0.03	0.014
			0.75	0.03	0.023

\*Landolt et al. (1985) also provided consumption data by location for particular species. In areas where consumption of a fish species (by all populations combined) exceeded the highest consumption rate of that species by a particular population, the consumption rate of that species and the contaminant level in that species at that location are provided. This was the case for Pacific hake, starry flounder and Pacific cod.

\*\* Consumption rates are expressed as geometric mean grams of cleaned fish available for consumption per person per day during time period each species was present in fishery. However, estimated mercury intake levels are based on 12 month consumption.



**Table 5.** Puget Sound Boat Angler Data (Landolt et al., 1987). Estimated mercury intake levels for populations consuming particular species at two Puget Sound locations (both industrialized areas).

Fish	Location in Puget Sound	Consumption rate* (g/kg/day)	Fish tissue Hg concentration (mg Hg/kg fish)	Estimated Hg intake (µg/kg/day)
Rock Sole	Commencement Bay (C.B.)	0.098	0.003	0.0003
Walleye-Pollock	C. B.	0.022	0.016	0.0004
Pacific Hake	C.B.	0.112	0.005	0.0006
Sablefish	Elliot Bay	0.495	0.013	0.0064
Pacific Cod	C. B.	0.315	0.030	0.0095

\* Consumption rates are expressed as geometric mean grams of cleaned fish available for consumption per person per day during time period each species was present in fishery. However, intake levels presented are based on 12 month consumption.

**Table 6.** Estimated mercury intake determined from Tulalip Tribes, Squaxin Island tribe and Suquamish tribe consumption data combined with contaminant Washington State Department of Fish and Wildlife data.

	<b>Finfish Group Consumed</b>	<b>N</b>	<b>Consumption in g/kg/day (90th% pop. values)</b>	<b>Fish mercury level median of composite means in mg Hg/kg fish</b>	<b>Intake in µg Hg/kg/day **</b>	<b>Consumption in g/kg/day to obtain 0.08 µg Hg/kg/day (% of pop.)***</b>
<b>Tulalip Tribes</b>	Anadromous	73	1.429	0.1 (Chinook Salmon)	0.14	0.8 (86%)
				0.05 (Coho Salmon)	0.07	1.6 (92%)
	Anadromous	73	0.63 (86th%)*	0.1 (Chinook Salmon)	0.06	0.8 (86%)
				0.05 (Coho Salmon)	0.03	1.6 (92%)
	Pelagic	73	0.156	0.29 (Quillback Rockfish)	0.05	0.28 (100%)
<b>Squaxin Is. Tribe</b>				0.17 (Copper Rockfish)	0.03	0.47 (100%)
	Bottom	73	0.111	0.06 (English Sole)	0.01	1.3 (100%)
	Anadromous	117	1.639	0.1 (Chinook Salmon)	0.16	0.8 (75%)
				0.05 (Coho Salmon)	0.08	1.6 (90%)
	Pelagic	117	0.106	0.29 (Quillback Rockfish)	0.03	0.3 (96%)
<b>Suquamish Tribe</b>				0.17 (Copper Rockfish)	0.02	0.5 (100%)
	Bottom	117	0.176	0.06 (English Sole)	0.01	1.3 (100%)
	Salmon	92	1.680	0.1 (Chinook salmon)	0.17	
				0.05 (Coho salmon)	0.08	
	Halibut/Sole/Rockfish/Flounder/Red Snapper	76	0.392	0.29 (Quillback rockfish)	0.11	
				0.17 (Copper rockfish)	0.07	
				0.06 (English sole)	0.02	
	Tuna	83	0.346	0.17 (Canned tuna)	0.06	

\* Consumption value based on 86<sup>th</sup> percentile consumption rate for anadromous fish category.

\*\* These intake values are based on the assumption that the fish type consumed from a particular group (anadromous, pelagic, bottom) is of one type only (provided in fifth column).

\*\*\* Last column indicates consumption allowed (for species listed in column four) to remain under upper bound of tolerable daily intake (values could not be determined from Suquamish tribe data).